Magnetic field–switchable millimeter wave switch for 81, 94, and 140 GHz based on metal substituted ε-iron oxide

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§ 1. Methods

Materials. A series of ε-$Ga_xFe_{2-x}O_3$ and ε-$Al_xFe_{2-x}O_3$ were obtained by the sol–gel method according to the previous study.[38] An aqueous ammonia solution was added into a mixed aqueous solution of iron nitrate and gallium nitrate (or aluminum nitrate) while stirring. Then tetraethyl orthosilicate (TEOS) was added to form a silica matrix. The precipitation was filtered, washed with water, and dried, yielding the precursor. Using an electric furnace, the precursors were sintered in air for 4 hours at 1000−1100 ºC. Finally, a sodium hydroxide aqueous solution was added to the sintered sample and stirred for one day at 60 ºC. Finally, the product was collected by filtration, washed with water, and dried.
Physical Property Measurements. Elemental analyses were performed with XRF using ZSX Primus IV/NT (RIGAKU). The PXRD patterns were measured using Ultima IV (Rigaku) with Cu Kα radiation (λ = 1.5418 Å), and the Rietveld analyses of the XRD patterns were conducted with PXDL software (RIGAKU). TEM images were acquired using JEM 2000EX (JEOL). The magnetic hystereses were measured using a superconducting quantum interference device magnetometer (SQUID, MPMS 7, Quantum Design).
Millimeter Wave Absorption Measurements. The millimeter wave absorption properties were measured with a THz-TDS system, TAS7400 (Advantest). A THz light was generated with a GaAs photoconductive antenna and a 1550-nm fiber laser. The generated THz light was condensed with a silicon lens and paraboloidal mirrors. Then it was irradiated to the pellet samples. The transmitted THz light was detected with a GaAs photoconductive antenna, and the electric fields of the transmitted THz light wave were obtained in the time domain. The absorption spectra were acquired by Fourier transformation. Powder samples were compressed into 13 mmφ pellets with thicknesses of 1.779 mm (**1**), 2.322 mm (**2**), 2.145 mm (**3**), 2.374 nm (**4**), 2.199 mm (**5**), and 2.169 mm (**6**).The filling ratios were 60.2 vol.% (**1**), 59.6 vol.% (**2**), 58.4 vol.% (**3**), 51.0 vol.% (**4**), 51.4 vol.% (**5**), and 57.1 vol.% (**6**).

§ 2. TEM images

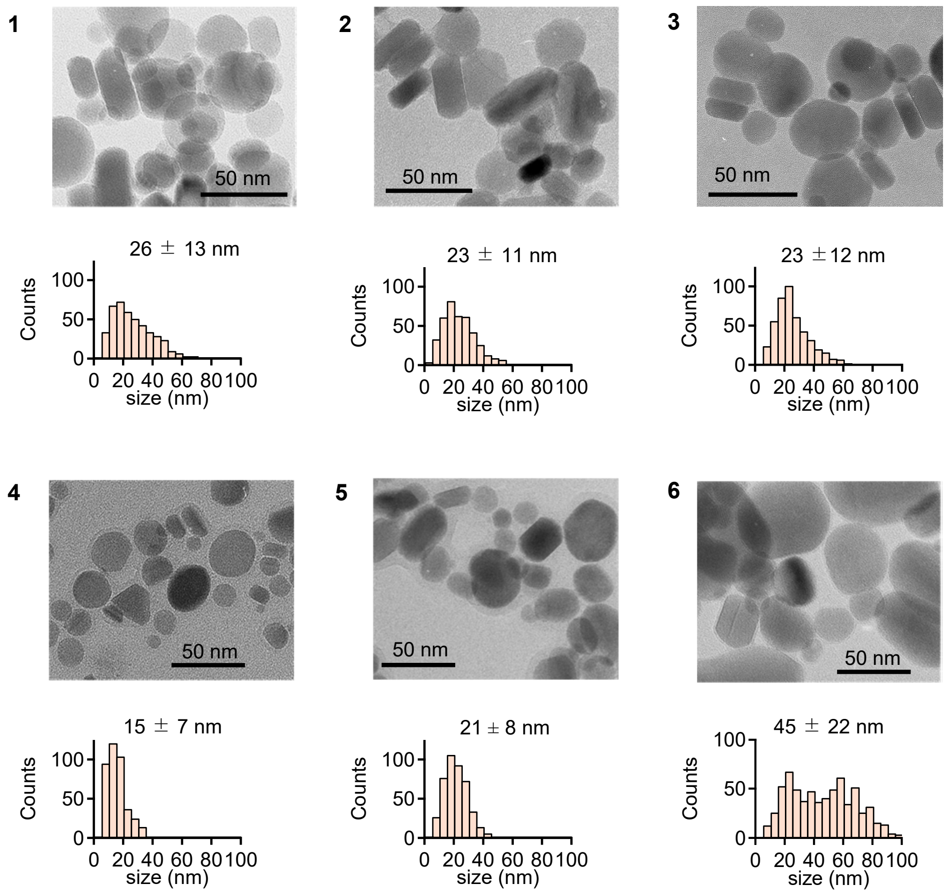

Figure S1. TEM images with size distribution.

§ 3. PXRD patterns with Rietveld analyses

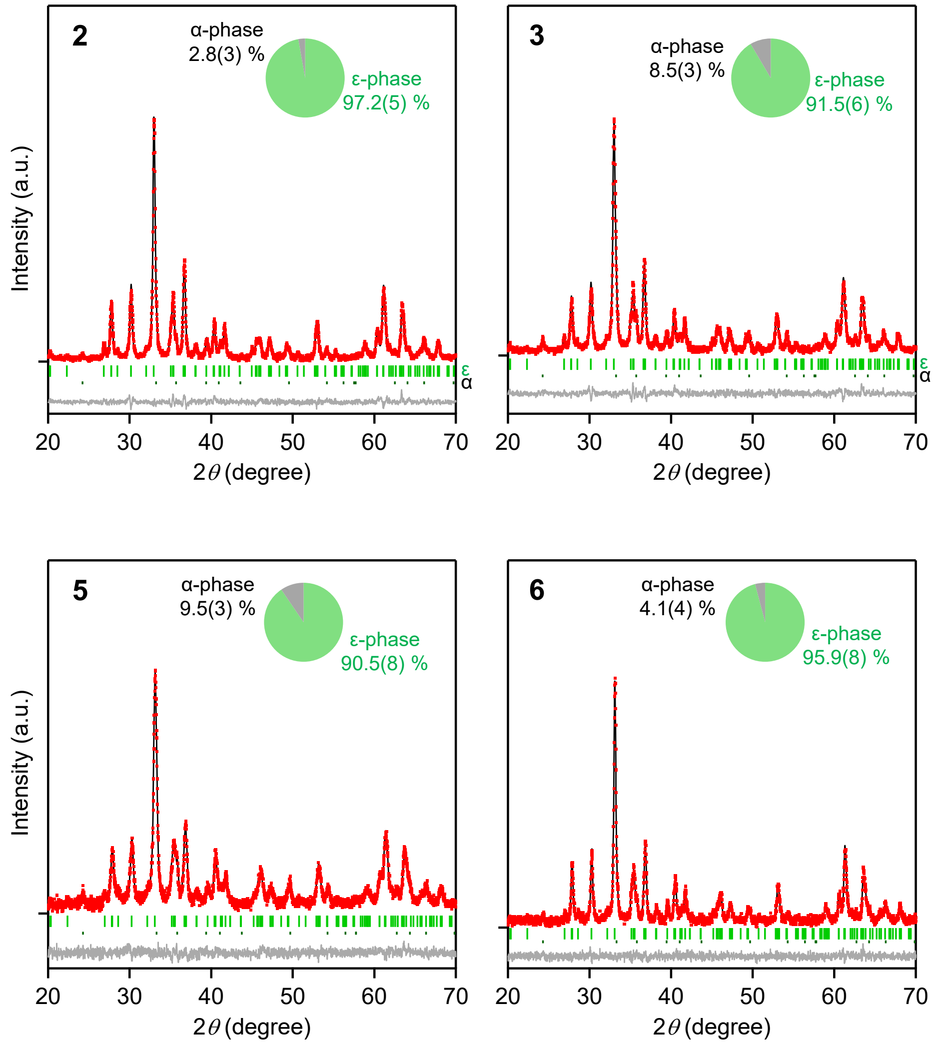

Figure S2. PXRD patterns and Rietveld analyses for **2, 3, 5** and **6**. Red dots, black lines, and gray lines are the observed, calculated, and their residual patterns, respectively. Green and black bars represent the calculated Bragg reflection positions of the ε- and α-phases, respectively.

§ 4. Millimeter wave absorption spectrum analyses

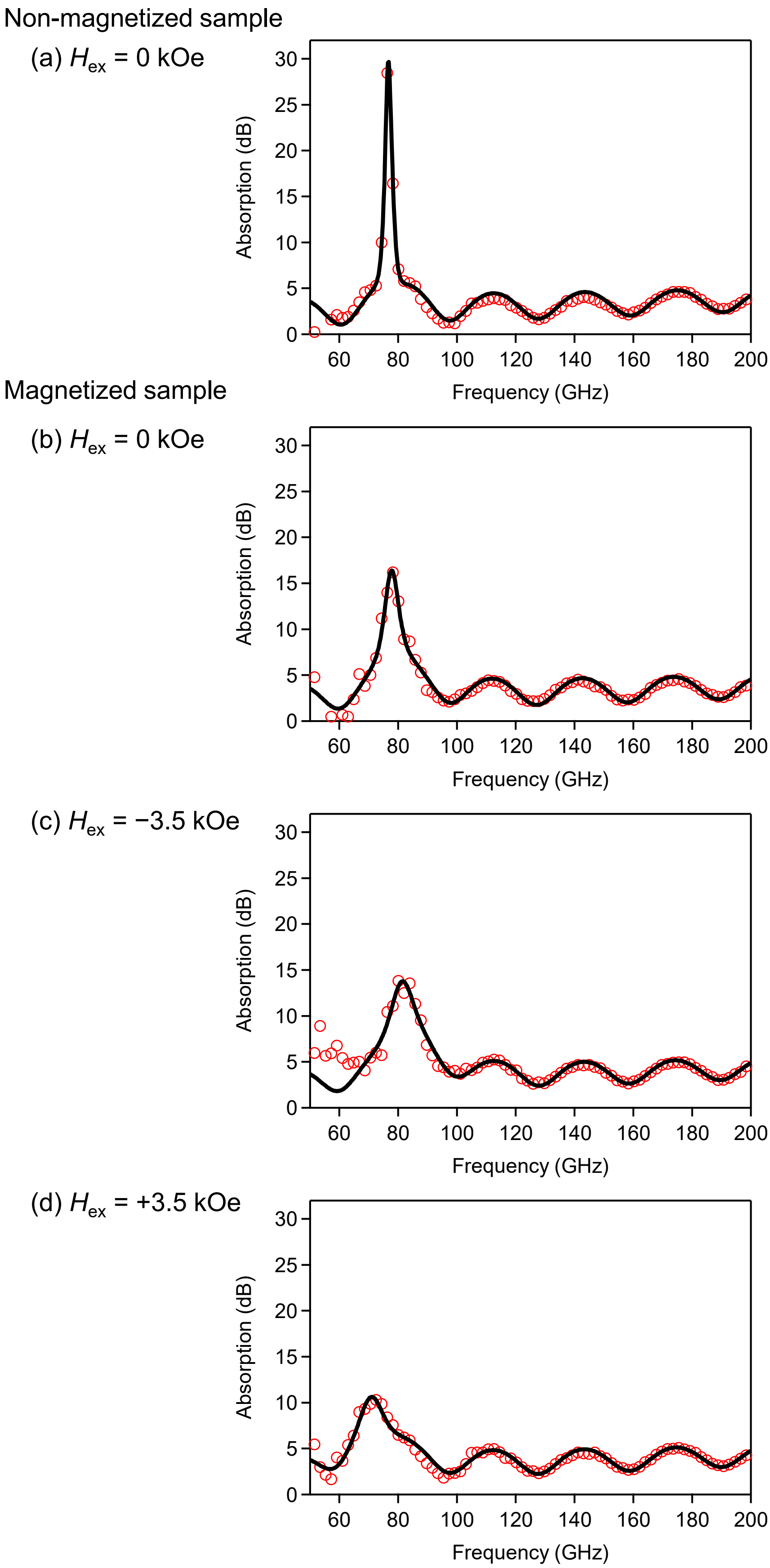

Figure S3-1. Millimeter wave absorption spectra for **1** measured with a non-magnetized sample under (a) H_{ex} = 0, a magnetized sample under (b) H_{ex} = 0, (c) H_{ex} = −3.5 kOe, and (d) H_{ex} = +3.5 kOe. The open circles represent the observed spectrum and the black lines represent the fitted spectrum.

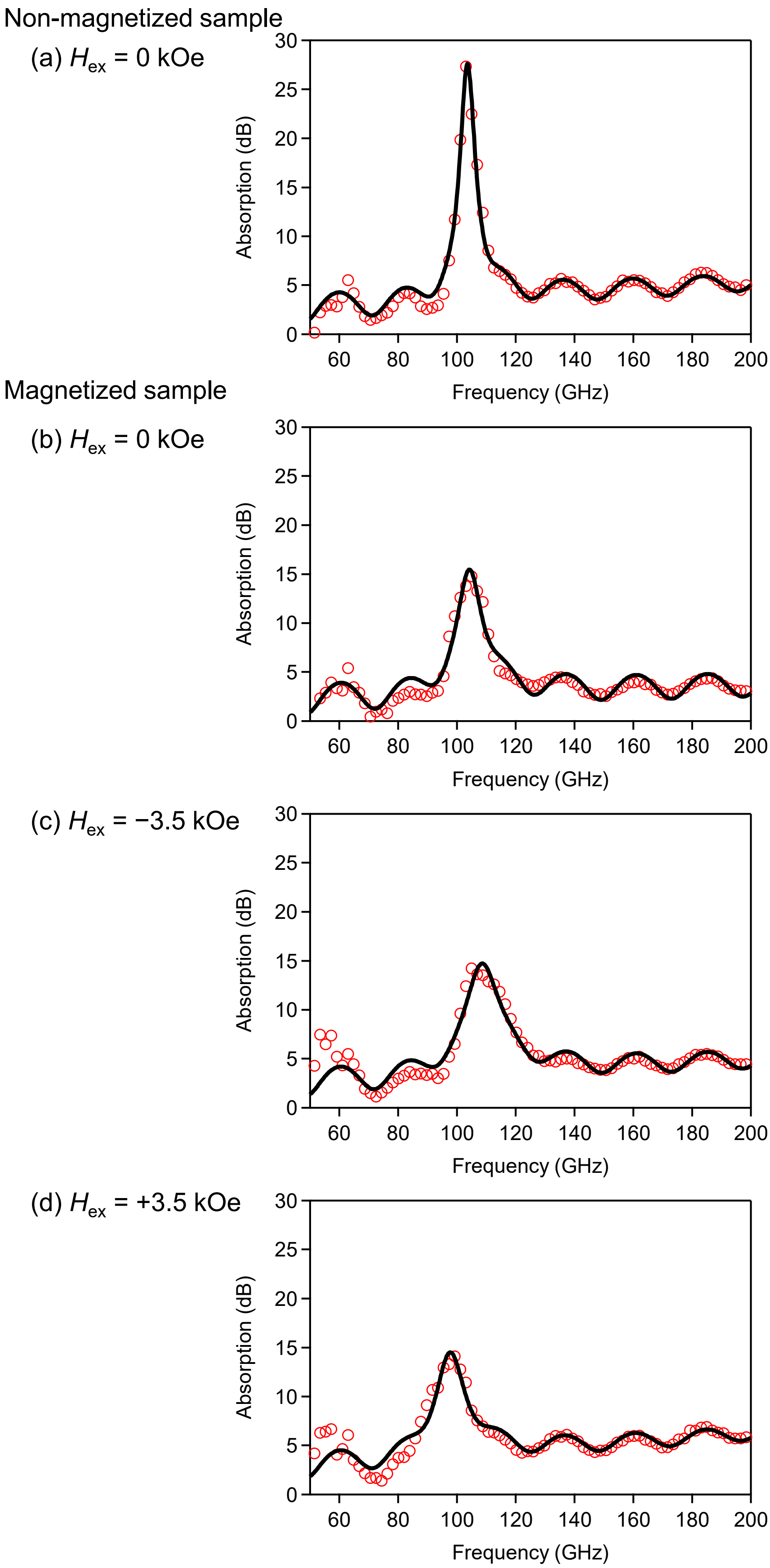

Figure S3-2. Millimeter wave absorption spectra for **2** measured with a non-magnetized sample under (a) H_{ex} = 0, a magnetized sample under (b) H_{ex} = 0, (c) H_{ex} = −3.5 kOe, and (d) H_{ex} = +3.5 kOe. The open circles represent the observed spectrum and the black lines represent the fitted spectrum.

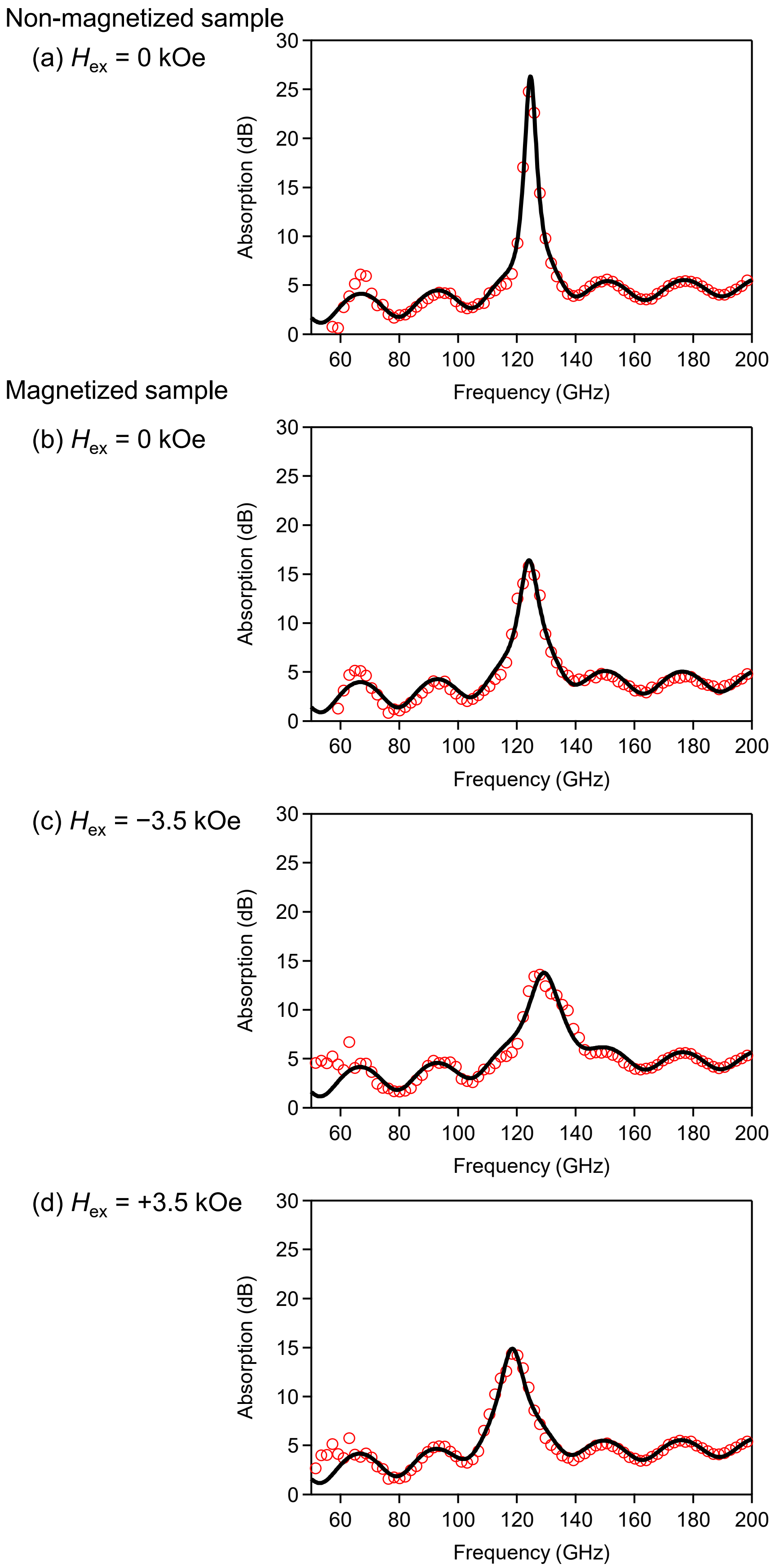

Figure S3-3. Millimeter wave absorption spectra for **3** measured with a non-magnetized sample under (a) $H_{ex} = 0$, a magnetized sample under (b) $H_{ex} = 0$, (c) $H_{ex} = -3.5$ kOe, and (d) $H_{ex} = +3.5$ kOe. The open circles represent the observed spectrum and the black lines represent the fitted spectrum.

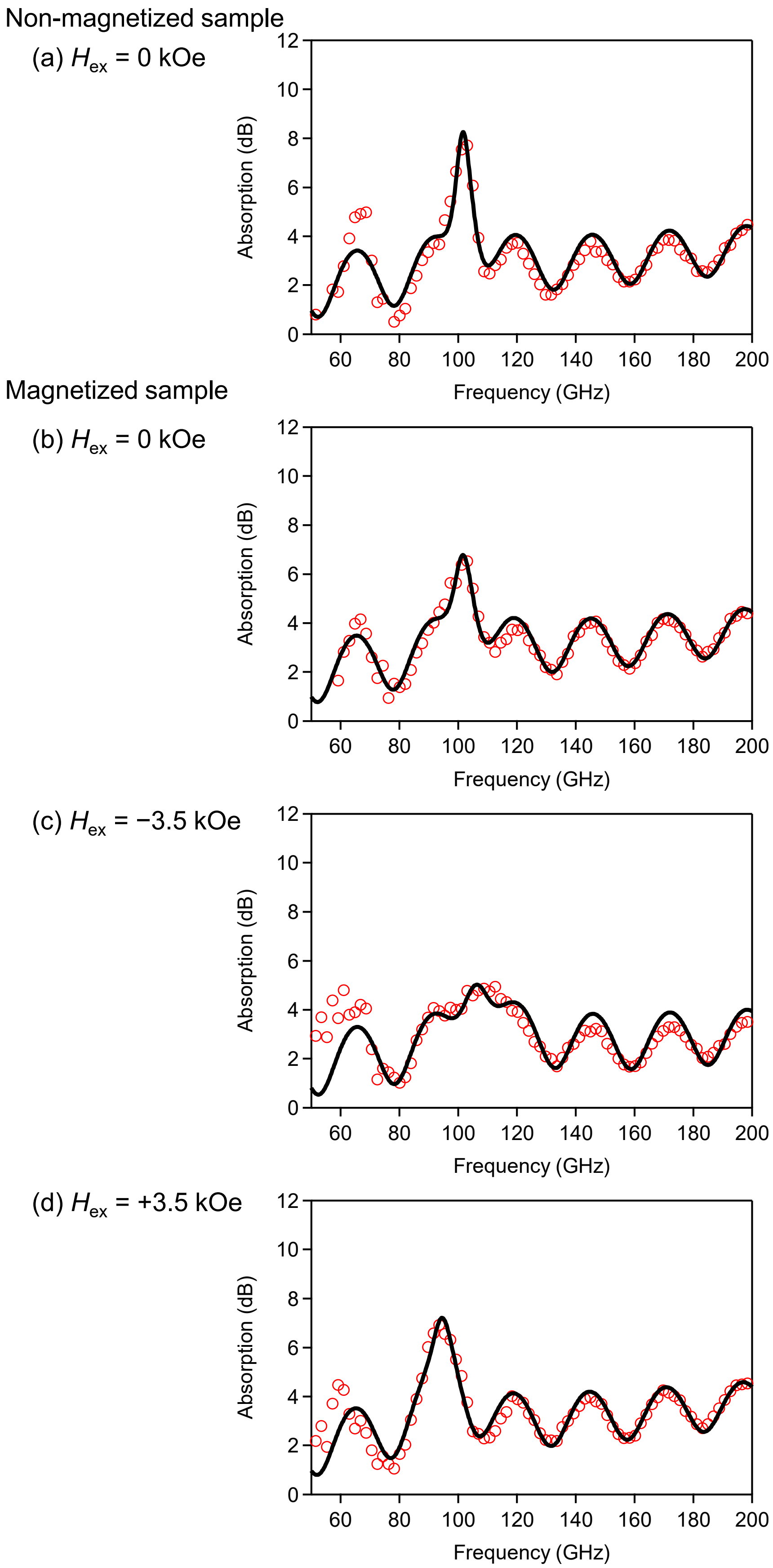

Figure S3-4. Millimeter wave absorption spectra for **4** measured with a non-magnetized sample under (a) $H_{ex} = 0$, a magnetized sample under (b) $H_{ex} = 0$, (c) $H_{ex} = -3.5$ kOe, and (d) $H_{ex} = +3.5$ kOe. The open circles represent the observed spectrum and the black lines represent the fitted spectrum.

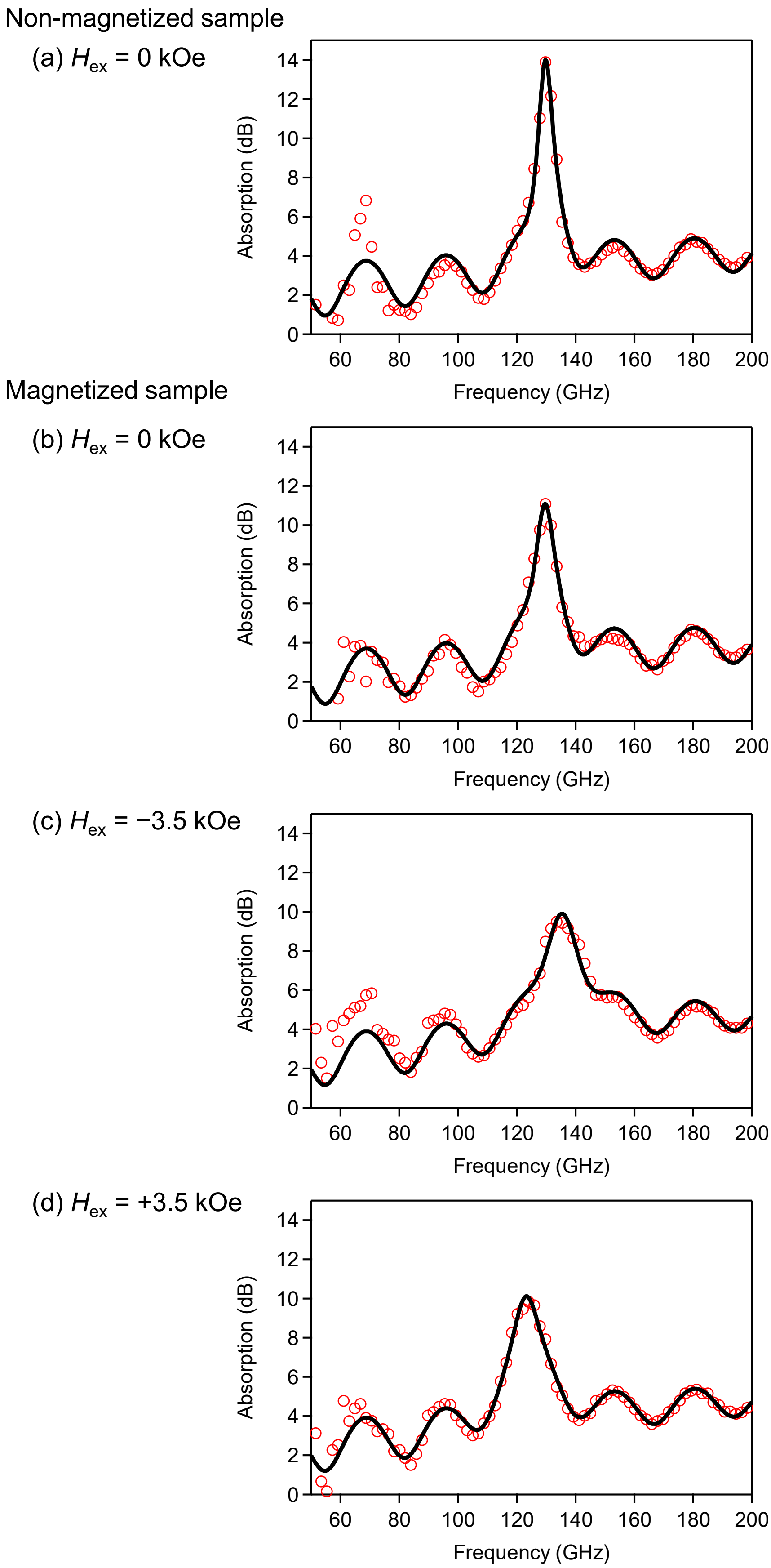

Figure S3-5. Millimeter wave absorption spectra for **5** measured with a non-magnetized sample under (a) $H_{ex} = 0$, a magnetized sample under (b) $H_{ex} = 0$, (c) $H_{ex} = -3.5$ kOe, and (d) $H_{ex} = +3.5$ kOe. The open circles represent the observed spectrum and the black lines represent the fitted spectrum.

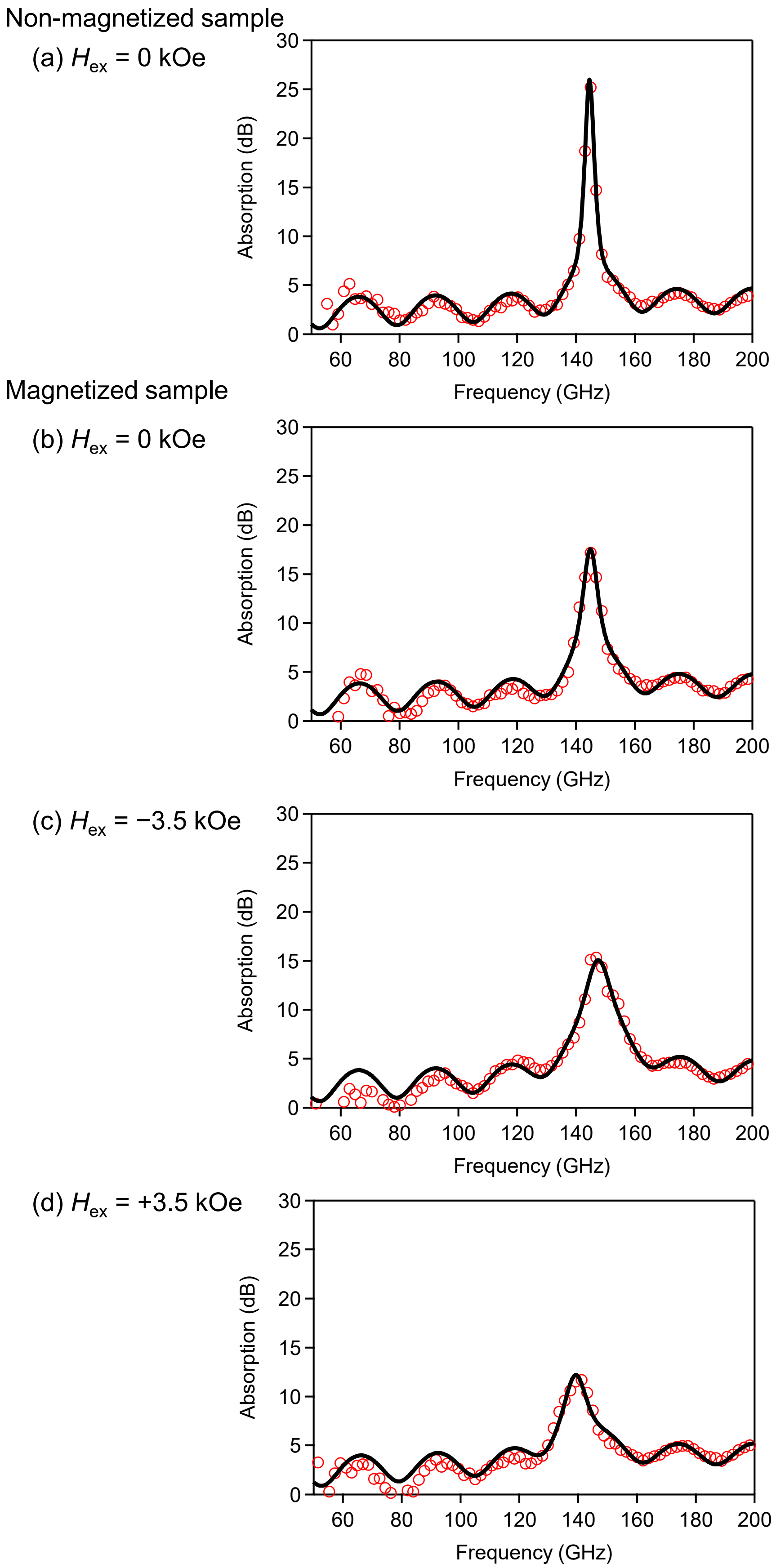

Figure S3-6. Millimeter wave absorption spectra for **6** measured with a non-magnetized sample under (a) $H_{ex} = 0$, a magnetized sample under (b) $H_{ex} = 0$, (c) $H_{ex} = -3.5$ kOe, and (d) $H_{ex} = +3.5$ kOe. The open circles represent the observed spectrum and the black lines represent the fitted spectrum.

§ 5. Landau-Lifshitz-Gilbert model calculation.

The dynamics of $\boldsymbol{M}$ were obtained by varying the frequency of EM wave (f). The value of $1-M_z$ (M_z is the z-component of magnetization) at a steady state was calculated to obtain the absorption spectrum. In the experiment, linearly polarized THz light was irradiated onto the sample and can be expressed as the sum of right and left circularly polarized light. The magnetization precession is only induced by the right (left) circularly polarized light when the magnetization is parallel (+direction) [antiparallel (–direction)] to the direction of the incident electromagnetic wave.

The following was used to consider such a selection rule in the Landau-Lifshitz-Gilbert model calculations. First, we calculated the dynamics of magnetizations with 5,000 random directions and plotted the average of $1-M_z$ vs. f. Then we assumed that right (left) circularly polarized light was only absorbed in the case of the +direction (–direction) with a transmittance of 0.998 per direction. In other words, when the numbers of magnetizations in the +direction and the –direction were x and $5{,}000-x$, respectively, the absorption coefficient was expressed as $1-0.5\times(0.998^{x}+0.998^{5000-x})$. The overall absorption property was calculated by applying this coefficient to the above $1-M_z$ vs. f plot.

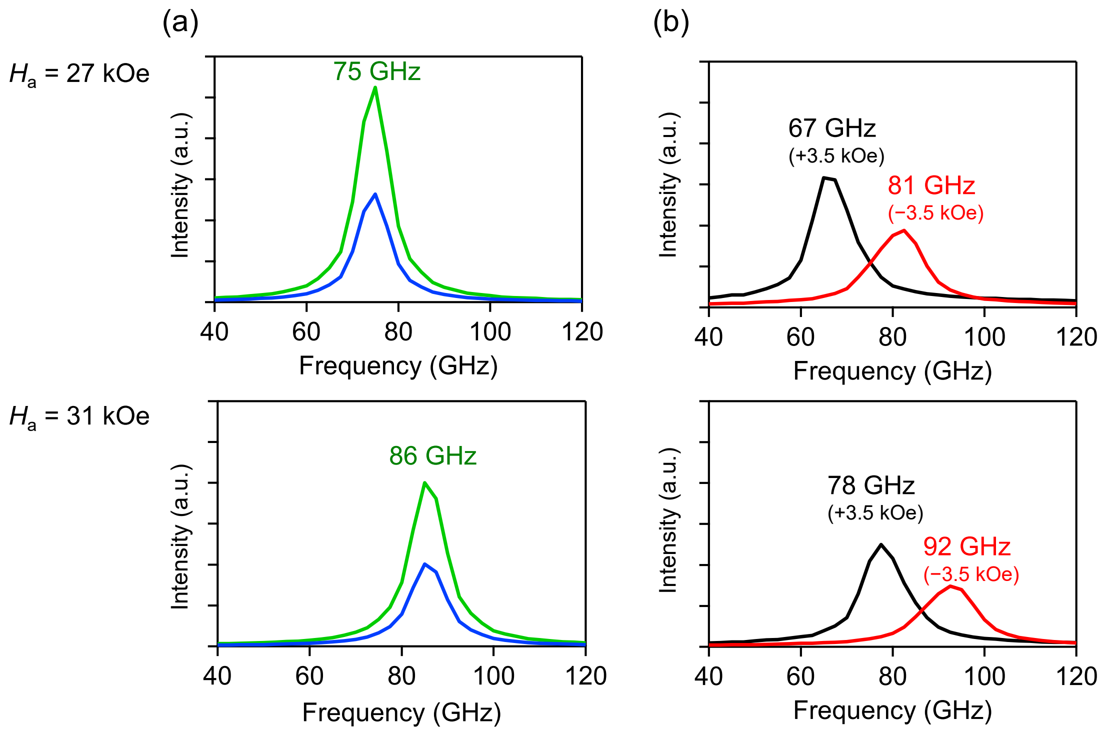

Figure S4. Landau-Lifshitz-Gilbert model calculation for H_a = 27 kOe and 31 kOe. (a) Calculation for the as-prepared pellet-form sample (green) and magnetized sample (blue) under H_{ex} = 0. (b) Calculation for the magnetized pellet sample under H_{ex} = +3.5 kOe (black) and −3.5 kOe (red).